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Operation of a Proportional Counter at Low Temperature for Applying to CEMS

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Operational technique of a gas-filled proportional counter was investigated at low temperatures for the purpose of applications to the conversion electron Mössbauer spectroscopy (CEMS). The proportional counter works well and can be applied to CEMS at low temperature if the counter is filled with purified helium at temperature below 22K, with purified neon at 22–53K, and with a gas mixture of He+5%N_2 or He+10%CO at temperature higher than 46 or 47K, respectively. However, the pure neon is not suitable for CEMS measurements because of high non-resonance counting efficiency for X and γ rays. If the counter is filled with a gas mixture of helium and neon, more efficient CEMS measurements may be possible at the temperature range of pure neon.

KEYWORDS : CEMS/Proportional counter/Low temperature/Mössbauer spectroscopy/

I. INTRODUCTION

Proportional counters have been used in many experimental fields because of their comparatively high energy resolution, of good counting efficiency for low-energy radiations and of variety of fabrication for experimental purposes. Until quite recently, however, the operation of a proportional counter has been little investigated at low temperature, especially below 77K, which restricts the applications of the counter. Several years ago, Isozumi *et al.* established the technique for operating stably a proportional counter, by filling with pure helium gas, at temperature below 5K [1–4]. This technique has given a new experimental way and applied to the conversion electron Mössbauer spectroscopy (CEMS) for studying the magnetic properties of corrosion products at the surface of iron [5,6]. It has been also expected to operate a counter at temperature between 5–77K because it is usually desired to do experiments at various temperatures. We have therefore recently investigated the operation of the counter in this temperature range [7–9]. In the present report, our recent studies on the operation of a proportional counter at low temperature are summerized and the practical applications to CEMS were described.

II. OPERATION OF A PROPORTIONAL COUNTER

The cryostat and counter assembly are shown in Fig. 1. The proportional counter is installed in the counter room which is filled with counter gas of 1 atm at room temperature. The heat is

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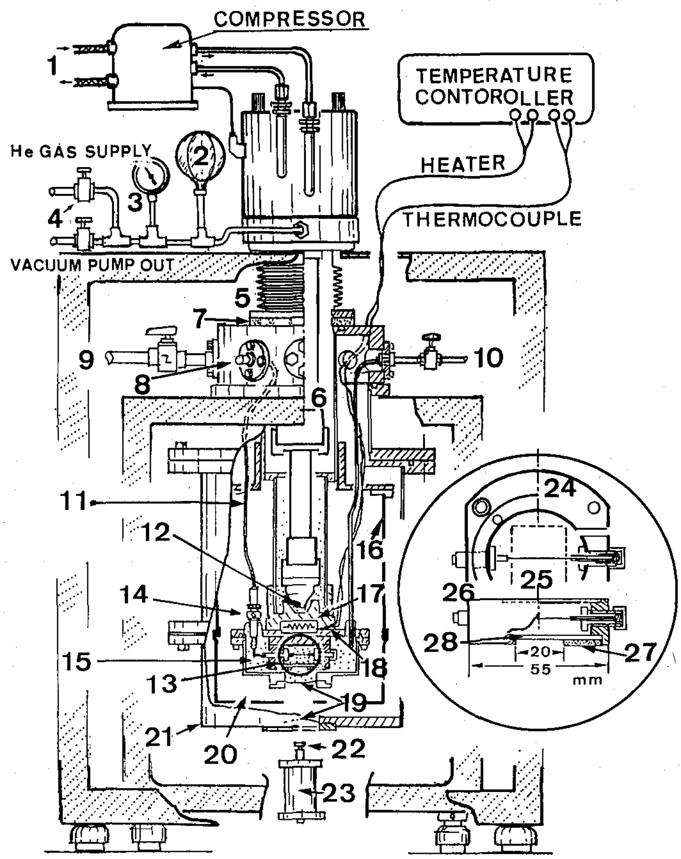

Fig. 1. Cryostat and counter assembly. 1 : Cooling water for the compressor; 2 : pressure buffer baloon; 3 : pressure gauge; 4 : valve for heat exchanging helium gas; 5 :bellows (vibration absorber); 6 : expansion engine; 7 : insulation flange; 8 : connector for signals and high voltage bias; 9 : vacuum valve; 10 : counter gas inlet; 11 : 5-mm-diameter SUS tube for signal and high voltage lead; 12 : heat exchange space filled with helium gas; 13 : proportional counter; 14 : SHV connector for high voltage bias; 15 : counter room; 16 : copper radiation shield; 17 and 18 : heater and thermocouple for controlling temperature; 19 : windows; 20 : vacuum space; 21 : outer vacuum jacket; 22 : ^{57}Co source for CEMS; 23 : velocity transducer for CEMS; 24 : counter body; 25 : anode wire (30-μm gold-coated tungsten wire); 26 : specimen (a sample foil for CEMS measurements or ^{57}Co source for examining the counter operation); 27 : collimator (2-mm-thick lead plate); 28 : absorber (3-mm-thick Lucite plate).

taken away by adiabadic expansion in the expansion engine of high-pressure helium gas introduced from the compressor. The counter can be kept at any temperature between 13.3–300K by controlling the heater current. Two fine thermocouples of Au+0.07%Fe-chromel are placed in the counter room for monitoring the temperature and near the heater for controlling the temperature. More detailed descriptions of the cryostat and counter assembly are given elsewhere [7].

For examining the counter operation, a ^{57}Co source ($\sim$1 kBq) electroplated on an aluminum foil was mounted on the cathode plate of the counter. Energy spectra of low-energy electrons emitted from the ^{57}Co source were recorded by filling purified helium, purified neon and gas mixtures, He+5%N_2 and He+10%CO, into the counter. The purified helium and neon are introduced into the counter through a trap of molecular sieves cooled with liquid nitrogen.

The counter signals due to low-energy electrons from the ^{57}Co source were distinctly separated from noise level at temperature below 22K if purified helium is filled, at temperature between 22–53K if purified neon is filled and at temperature above 46 or 47K if He+5%N_2 or He+10%CO is filled, respectively. The typical examples of the energy spectra are given in Figs. 2–4. The main peak in the spectra is caused from superimposed 5.6-keV K-LL Auger electrons and 7.2-keV K-conversion electrons and the coincidence detections of these electrons make the other small peak corresponding to 12.8 keV.

Most of the impurity molecules in helium is frozen at very low temperature and the counter works with a mechanism different from that for a counter filled with a gas mixture [2]. The impurity molecules evaporate at higher temperature and eventually block the working of the counter at some temperature around 23K.

Liquefaction of neon makes the operation of the counter filled with purified neon unstable at temperature below 21K. At temperature higher than 53K, the operational instability of the counter occurs again due to the evaporation of impurity molecules.

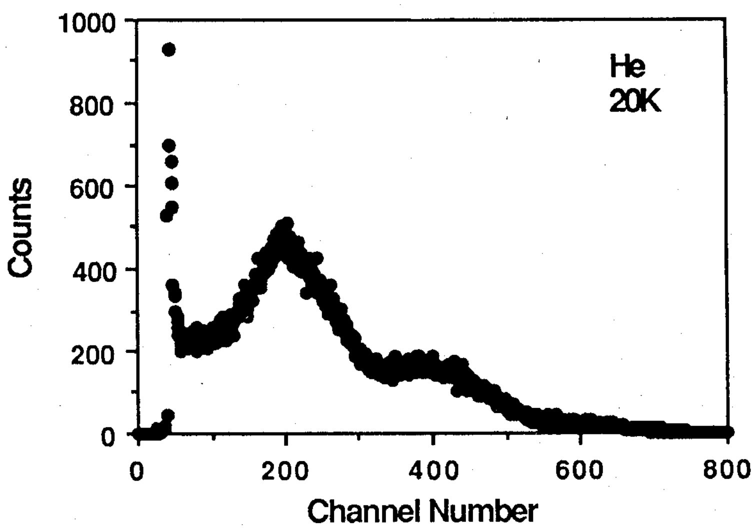

Fig. 2 Energy spectrum of electrons emitted from the ^{57}Co source at 20K by filling purified helium in the counter.

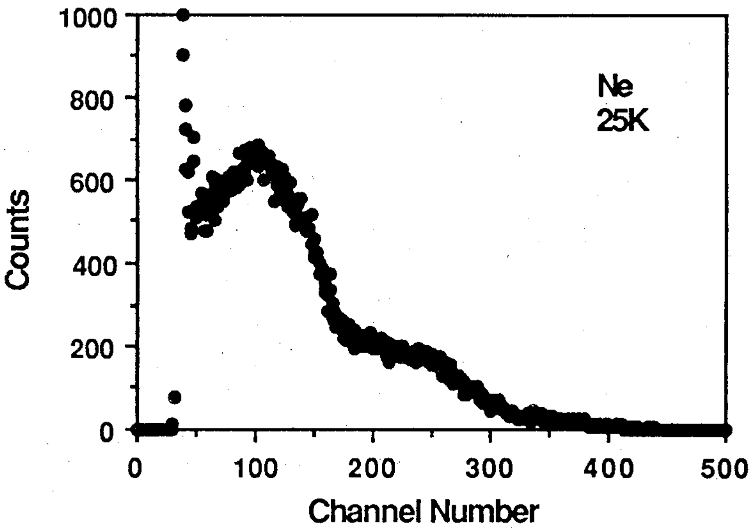

Fig. 3. Energy spectrum of electrons emitted from the ^{57}Co source at 25K by filling purified neon in the counter.

When the counter is filled with the gas mixture, it works with the well known mechanism. Partial liquefaction of N_2 or CO causes the decreasing of the operational bias voltage of the counter at low temperature, lower than 53K for He+5%N_2 or 59K for He+10%CO, respectively, and at temperature lower than 46 or 47K the counter becomes unstable. More detailed discussions are described in our previous literatures [7,8].

III. APPLICATIONS TO CEMS

In order to examine the counter system performance for CEMS studies, CEMS spectra of a natural iron foil mounted on the cathode plate were observed with a ^{57}Co source in an Rh matrix (1.85 GBq) at several temperatures by filling the above-mentioned gases and gas mixtures into the counter. The obtained spectra are shown in Fig. 5. The relative resonance peak-heights with pure helium and gas mixtures are 7–8%, but the peak-heights are conspicuously low ($\sim$3%) in the cases of pure neon gas.

Figure 6 shows the energy spectra obtained at 25K by filling neon in the counter with the Mössbauer source placed outside the counter, in the CEMS conditions, and with the electroplated source mounted in the counter. Indistinct and distinct peaks in the pulse-height ranges (A) and (B), respectively, and a wide base in the range (C) are seen in the spectrum with the Mössbauer source. The peaks in the ranges (A) and (B) are probably caused, respectively, by the 6.4-keV K-X rays and 14.4-keV γ rays from the source. The wide base in the range (C) may be due to the 122-keV γ rays. The peaks in the spectrum with the electroplated source are due to the Auger and conversion electrons as described before. On the contrary, in the cases of helium and gas mixtures, no definite peak or wide base is found in the spectrum with the Mössbauer source owing

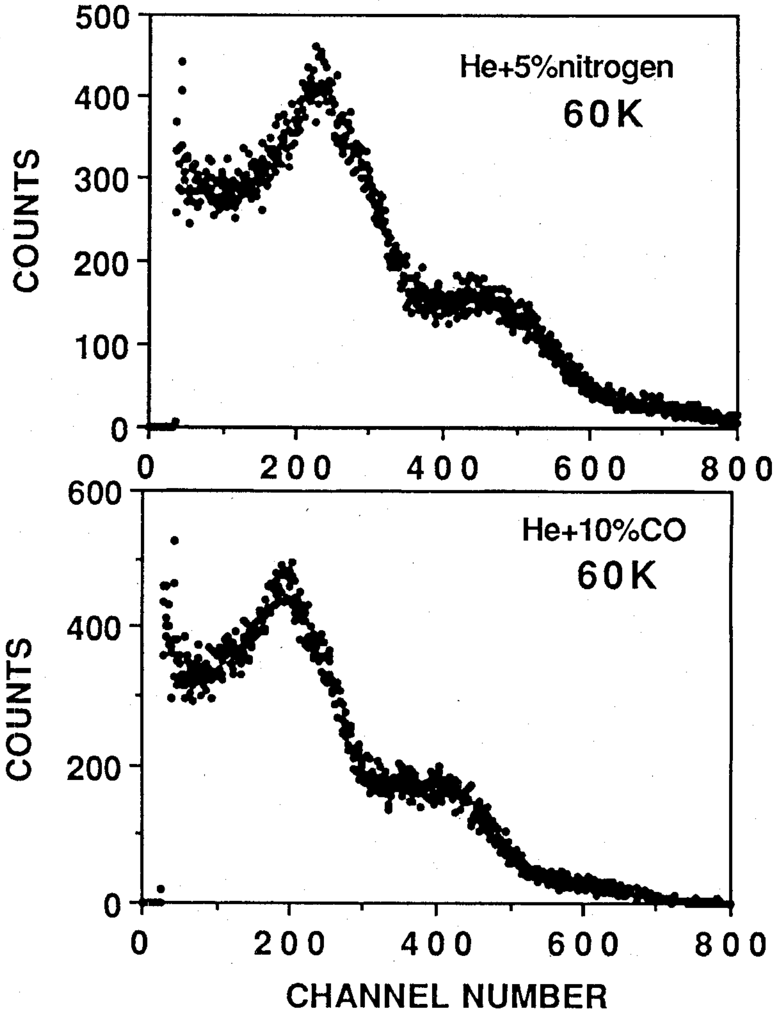

Fig. 4. Energy spectra of electrons emitted from the ^{57}Co source at 60K by filling $He+5\%N_2$ and $He+10\%CO$ in the counter.

to the difference in the atomic number of neon and helium, as shown in Fig. 7.

Considering these facts, the ratio of the Mössbauer resonance-peak counts to non-resonance counts is expected to increase with reducing neon gas in the counter. For this reason, the operation of the counter with a gas mixture of helium and neon was examined. It was found the counter works well with the He-Ne mixture in a temperature range similar to that for pure neon. Figure 8 shows the energy spectrum obtained at 25K by filling the 50%He+50%Ne mixture in the counter with the Mössbauer source placed outside the counter, where the peak due to 14.4-keV γ

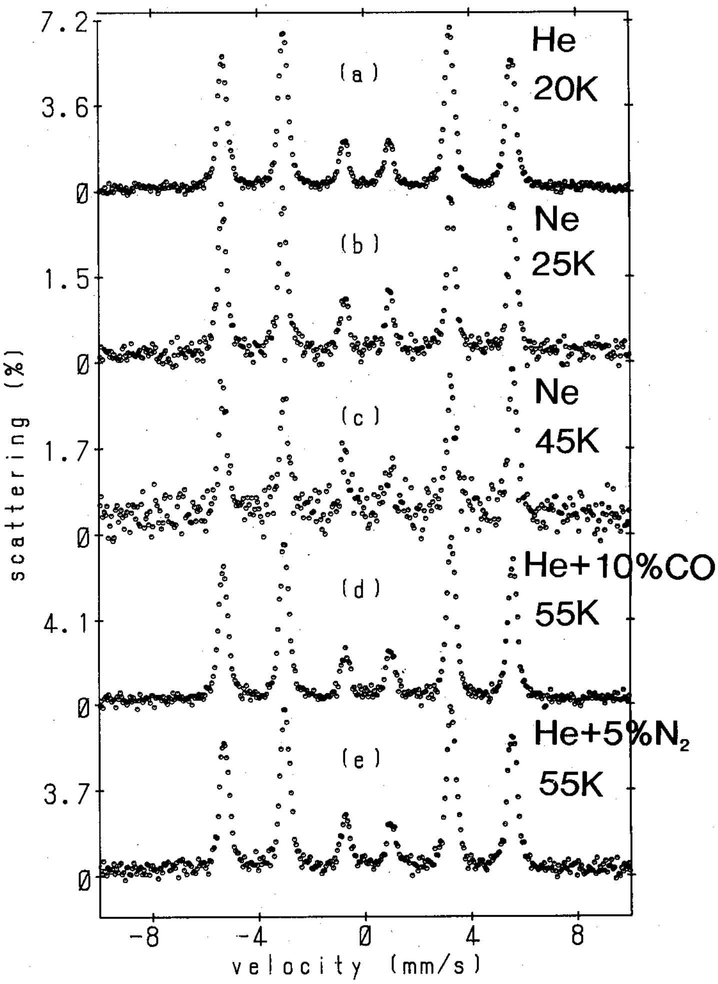

Fig. 5. CEMS spectra recorded at several temperatures. The counter gases are purified helium at 20K (a), purified neon at 25K (b) and 45K (c), and He+10%CO (d) or He+5%N_2 (e) at 55K.

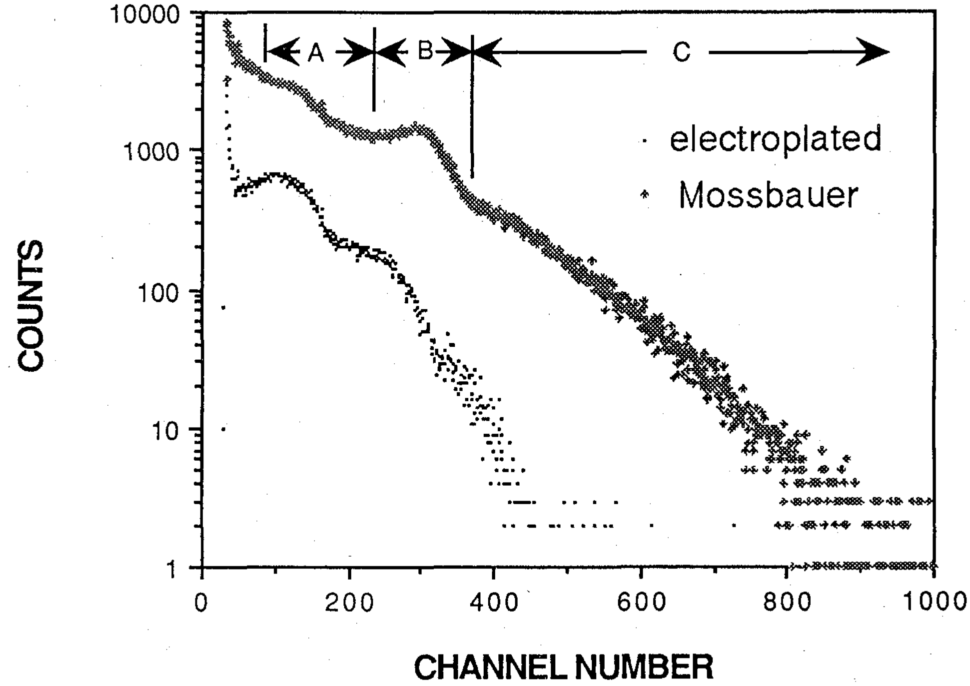

Fig. 6. Energy spectra observed at 25K by filling purified neon in the counter. One was recorded with the Mössbauer source placed outside the counter and the other with the electroplated source mounted in the counter.

rays is much reduced compared with the corresponding peak in Fig. 6. The conversion electrons emitted after the Mössbauer resonance absorption in the iron foil may be responsible mainly for the signals in the pulse-height range indicated by (A) in Fig. 6. Figure 9 shows the CEMS spectra of natural iron observed at 25K by using the counting signals in the ranges (A) and (B). The peak-height of approximately 6% obtained with signals in the range (A) is practically equivalent to that in the spectra of natural iron observed with pure helium or gas mixtures given in Fig. 5, while nothing but faint resonance peaks is found in the spectrum obtained with signals in the range (B). The peak-height in the CEMS spectra recorded with signals in the range (A) in Fig. 8 is given in Fig. 10 as a function of the partial pressure of neon in the He-Ne gas mixture where the total gas pressure is fixed to 1 atm. The peak-height increases with decreasing the neon pressure but does not increases more at pressures under 0.2 atm, on the other hand, the counting rate decreases with decreasing the neon pressure. We therefore consider the effective counting rate (ecr) defined as

$$ecr = \frac{peak\ height}{1 + peak\ height} \times (counting\ rate), \tag{1}$$

which gives a measure of experimental efficiency and is also shown in the figure. From Fig. 10,

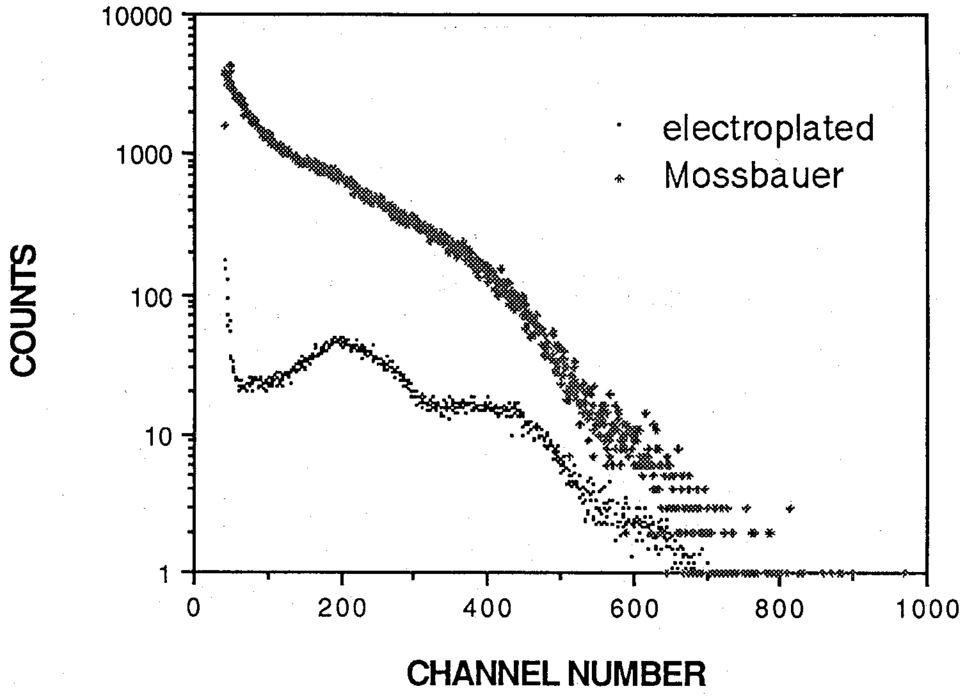

Fig. 7. Energy spectra observed at 20K with the counter filled with purified helium in the same conditions of the sources as in Fig. 6.

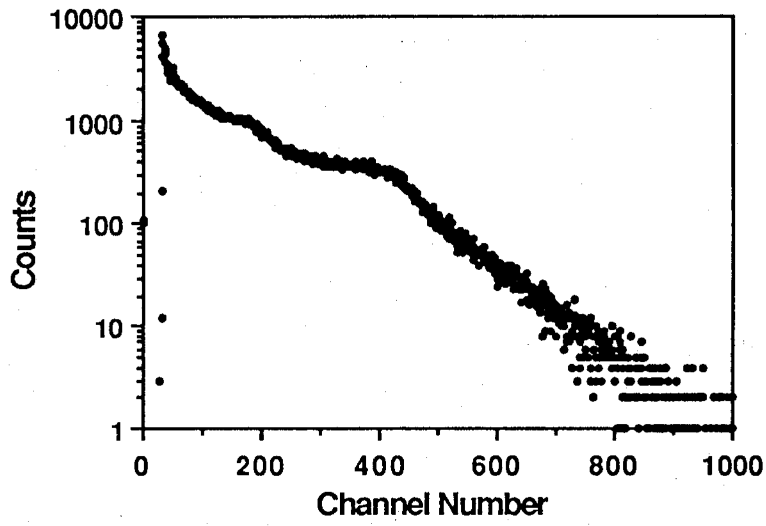

Fig. 8. Energy spectrum observed at 25K with the counter filled with 50%He+50%Ne mixture in the CEMS conditions.

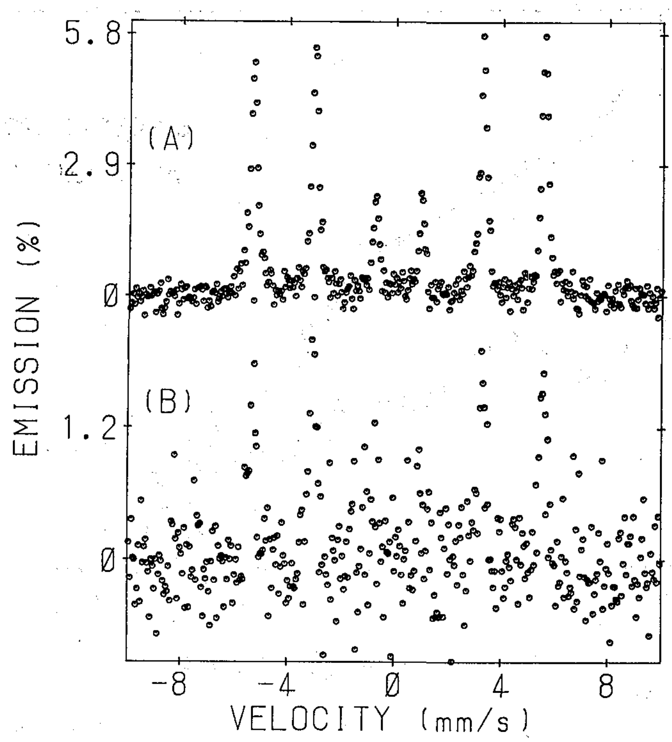

Fig. 9. CEMS spectra obtained by filling 50%He+50%Ne in the counter. The counter signals in the ranges (A) and (B) described in Fig. 8 are used for recording the respective spectra.

it can be concluded that a CEMS spectrum is obtained effectively at a partial pressure of neon between 0.2–0.5 atm if the total pressure of the He-Ne gas mixture is 1 atm. In this range of the neon pressure, the peak-height is also comparatively large, for instance, 7.5% when the partial pressure of neon is 0.2 atm. It was also confirmed that the counter works stably at 20–45 K by filling the 70%He+30%Ne mixture of 1 atm at room temperature.

It is easy to understand that the peak-height is large at a low partial pressure of neon because the counting efficiency of X and γ rays becomes small with decreasing the neon pressure. It is however difficult to elucidate that a proportional counter works well at 20–45K with a gas mixture of helium and neon in spite of instability of the counter filled with pure helium at temperature higher than 22K as descibed before.

In conclusion, it was confirmed that a gas-filled proportional counter can be applied effectively to CEMS at low temperature if the counter is operated by filling purified helium at temperature below 22K, by filling a gas mixture of helium and neon, for instance, 70%He+30%Ne at 20–45K, and by filling a gas mixture of He+5%N_2 or He+10%CO at temperature higher than 46 or 47K, respectively. Although the counter filled with purified neon works well at temperatures between 22–53K, the pure neon is not suitable for CEMS measurements because of high non-resonance counting efficiency for X and γ rays.

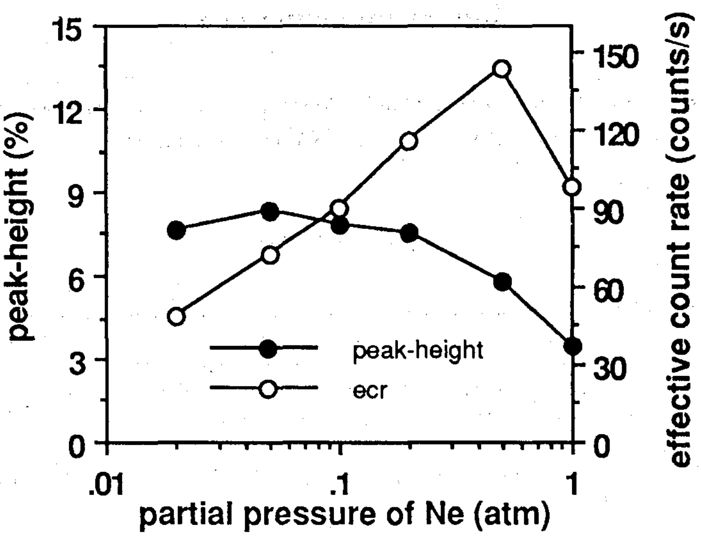

Fig. 10. Peak-height and effective counting rate as functions of the partial pressure of neon in the gas mixture where the total pressure is 1 atm.

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